Quantum theory of multiscale relaxation dynamics in molecular aggregates

Darius Abramavicius

[1]*Institute of Chemical Physics, vilnius University, Sauletekio al. 9-III, Vilnius, Lithuania*
Email: darius.abramavicius@ff.vu.lt

Photoinduced excitation transport and charge separation in in molecular systems has recently gained special attention due to high energy conversion efficiency in molecular solar cells. It has been observed that electrons and holes are very effectively generated on the inteface between two phases of the material even in very low externally applied electric fields. Early subpicosecond dynamics requires complete quantum description of such processes in order to reveal the underlying mechanisms of these processes. Stochastic Schroedinger Equation (SSE) approach has been applied to simulate charge coherent dynamics during primary charge separation events after photoexcitation process. The approach combines environment mediated statistical mixture of quantum propagation trajectories in the spirit of path-integral approach and allows to obtain distribution functions of physical variables. Dynamical processes of local heating and environment reorganization are revealed. Weak system-environment coupling regime demonstrates the classical results of charge hopping. We obtain two-fold charge separation kinetics: initial coherent charge separation is driven by quantum delocalization, what later evolves into diffusive hopping motion.